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The initial behavior of high altitude barium releases—I. The particulate ring

G. T. Best and H. S. Hoffman
Air Force Cambridge Research Laboratories, Bedford, Mass. 01730, U.S.A.

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Abstract—Large barium vapor releases at high altitude display an expanding particulate ring. Radiometrically calibrated photographic data from four releases has been analyzed and shows that the particulate cloud scatters light with a wavelength dependence of $\lambda^{-2\cdot5\pm1\cdot1}$, implying a very high proportion of particles of radius between 0·1 and 1·0 μ. The radial expansion velocity of the particles ranges from 0·1 to 0·7 km sec^{-1}, with a most probable value of 0·33 km sec^{-1}.

1. Introduction

The use of high altitude releases of barium vapor for the production of visible plasma clouds has been described by Foppl *et al.* (1965, 1967). There is considerable interest in maximizing the yield of ions per g of chemical carried, whether this be for use as a tracer for the extraterrestrial magnetic field, or for the production of dense clouds of ionization in the Earth's atmosphere. Numerous laboratory experiments have been performed by Brunner *et al.* (1969, 1970) and by Batalli-Cosmovici and Michel (1970, 1971) in order to optimize the yield of barium vapor, and the subsequent behavior of the vented barium vapor in the Earth's atmosphere has been described, in terms of the competing processes of oxidation and photoionization (Rosenberg *et al.*, 1968; Best and Rosenberg, 1969; Rosenberg and Best, 1971).

There are several possible methods of determining release efficiencies and vaporization processes from ground observations of high altitude releases. With suitable precautions and qualifications the total ion inventory made known by the scattered radiance can lead to an estimate of total vapor yield. Observations of the radial expansion of the vapor cloud can yield information through use of the snowplow model of Stuart (1965).

This note will consider the possibility that additional data concerning the release process can be determined from radiometric measurements of the particulate ring. Photographic data obtained during the vent phase of releases larger than 40 kg has shown that the non-vaporized component of the ejected material is also detectable, as a radially expanding ring which continues along the ballistic trajectory of the carrier rocket. In this note we discuss observations of a series of large releases conducted at Eglin AFB, Florida in January 1971. Figure 1 shows the ring formed by a 48 kg barium payload release at 150 km. It is known to be a ring rather than a shell since suitably located sites have viewed it edge-on as a straight bar-type configuration. It is known to be particulate since it scatters at every wavelength and even shows the more prominent Fraunhofer lines of the solar spectrum. It is also noted that its radial expansion velocity is less than that of the outer surface of the vapor cloud released at the same time, and it becomes

visible primarily because atmospheric drag brings the vapor cloud to a stop, whereas the particulate ring continues along the ballistic trajectory of the carrier rocket. This ring is not to be confused with the *ring-like* appearance of the spherical shell vapor cloud structure resulting from extra-atmospheric releases of barium vapor. The ring structure probably arises because the spin-stabilized rocket makes several revolutions during the vent period. Since filter photography data showed this ring structure for several releases in the light of several wavelengths it was decided to analyze the data, because the relative scattering efficiency as a function of wavelength could point to a particle size distribution, and the absolute radiance of the ring could possibly indicate the total mass of particulates present. Since other measurements both in the laboratory (Brunner *et al.*, 1970) and of releases (Best and Rosenberg, 1969) have indicated that only about 20 per cent of the excess barium was vaporized it may be expected that the remaining barium, and the other products of the copper oxide–barium thermite reaction, should be found in the particulate cloud. Several different sizes of releases were observed, but all were assembled from identical cannisters, each containing 16 kg of chemical mix, and it can be assumed that each vented independently of the others. The mix ratio was 2·5 Ba to 1 CuO, with 1·8 % barium azide added. For the sizes of the rings observed and the maximum possible masses involved, optical thickness cannot be a problem. A particle must be at least 0·1 μ in diameter to produce Mie scattering in the visible region, and 48 kgm of products ($1{\cdot}1 \times 10^4$ cm^3) spread out 0·1 μ thick would only present an area of 0·11 km^2. In the measurements reported here the minimum area of the ring was 8 km^2, since observations of the ring can only be made after the ambient atmosphere has retarded the (initially faster) leading edge of the vapor component of the release.

2. Data Reduction

The basic data consisted of radiometrically calibrated photographic images, taken through interference filters, at speeds of up to 5 frames sec^{-1}. In the case of the event at 150 km altitude, the relatively higher atmospheric density resulted in an early separation of the ring and the vapor cloud. In the other events at 185 and 250 km, the radial velocity of the vapor cloud persisted, and the background formed by the leading edge of this vapor cloud had to be subtracted to obtain intensity data relating to the ring alone. Due to the atmospheric drag on the vapor cloud, the center of the ring at later times did not coincide with the center of the vapor cloud, and indeed one half of the ring was frequently obscured by the vapor cloud. Therefore some judgment had to be exercised in the selection of the center point of the ring and in the background profile subtracted, but since a large number of frames from a wide selection of sites, events and wavelengths were analyzed, it was felt that these random errors would be reduced in significance. The correctness of this assumption is shown by the fact that at any wavelength, the extremes of data did not depart by more than a factor of two from the mean. A systematic increase of scattering efficiency with decreasing wavelength was derived. Range data from each of the two photographic sites concerned, to the release centers, were obtained from triangulation data provided by Dr. T. N. Davis of the University of Alaska.

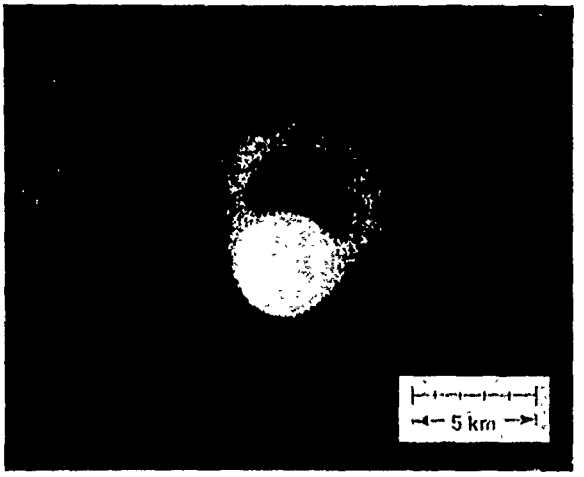

Fig. 1. Vapor cloud and particulate ring due to a 48 kg release at 150 km altitude, photographed at 6·5 sec after release through a 6142 Å filter. At this time and altitude the vapor cloud has been brought to rest and its radial expansion has already slowed.

3. Results

(a) *Radial expansion velocity*

Figure 2 shows the radial distribution of spectral radiance $B(r)$ at 6142 Å above sky background, of the ring resulting from the 150 km release at several times after release. If we plot the radius R at peak radiance, versus exposure time, we find that the peak moved with a radial velocity of $0{\cdot}33 \pm 0{\cdot}02$ km sec^{-1}. If the velocity and particle size distribution is frozen for all the observation time, it can be shown that by plotting $R^2B(r)$ versus (r/R), a single profile should describe all curves. There is some scatter which is attributed to uncertainty in the relative background level. (The peak ring radiance in the last two exposures is only just greater than the sky radiance.) Nevertheless all normalized profiles showed the same shape, with half-amplitude points at 0·17 and 0·51 km sec^{-1}, and practically zero component with velocity $v > 0{\cdot}7$ km sec^{-1}. Since the observed maximum gas-phase radial expansion velocity in these releases is $1{\cdot}4 \pm 0{\cdot}2$ km sec^{-1}, (Hoffman and Best, 1974), it is apparent that both the vapor and particulate velocities are higher than those reported in the laboratory experiments of Brunner *et al.* (1969), probably due to both heat transfer and inter-phase friction.

(b) *Particulate inventory*

The scattered spectral radiance intensity $dB/d\Omega$ and the scattering cross section $d\sigma/d\Omega = 1/I_o(d\mathcal{B}/d\Omega)$, where I_o is the solar irradiance, are given at four wavelegths in Table 1, normalized to 1 kg of chemical payload and averaged at all the observations at each wavelength.

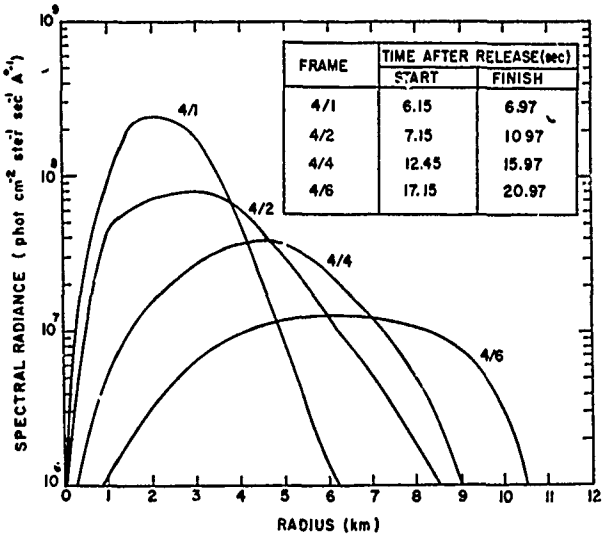

Fig. 2. Spectral radiance of particulate ring due to the release in Fig. 1, plotted as a function of radius for several times after release.

Table 1.

Wavelength (Å)	Spectral radiance $\frac{dB}{d\Omega}$ (phot. Å^{-1} sec^{-1} ster^{-1} kg^{-1})	Scattering cross section $\frac{d\sigma}{d\Omega}$ (cm^2 ster^{-1} kg^{-1})
4554	3·5 (18)	6·9 (4)
5350	3·6 (18)	6·8 (4)
5535	2·6 (18)	4·9 (4)
6142	1·8 (18)	3·2 (4)

The total scattering cross section falls by a factor of 2 from blue to red, indicating a fairly high proportion of smaller particles.

Batalli-Cosmovici and Michel (1970) have obtained in laboratory experiments a particle size distribution which may be represented by $dN(a) = Ca^{-k}\,da$ where dN is the number of particles of radius a within a size range of da. With the addition of 0·4% sodium azide, k increased from 2·15 to 2·75, *probably with a higher value of k for* $2\mu < a < 10\ \mu$. With this functional dependence of dN on a, the total volume of particulates present is dependent on the value of the upper radius limit in the integration for $k < 4$ and on the lower limit for $k > 4$, assuming that these limits differ by at least an order of magnitude. Thus there can be a considerable proportion of the total mass in a few large particles for $k < 4$, or in very small particle sizes for $k > 4$.

Let us next consider the integral of the scattering efficiency, Q_{sca}. The two observation sites subtend an angle of 15° at the cloud and the scattered light received does not appear to depend significantly on the scattering angle, which is close to 90° for both sites. Since the droplets are a homogeneous mixture of Ba, BaO and Cu in the molar ratio 3:2:2, we may treat the scattering as that due to conducting spheres, in which case (Van De Hulst, 1959, p. 163) the scattering is isotropic, except for the addition of diffractive scattering, which is only of consequence for small angles. We may write

$$4\pi \frac{d\sigma}{d\Omega} = \frac{4\pi}{I_o}\frac{dB}{d\Omega} = \pi a^2 \int_{a_{min}}^{a_{max}} Q_{sca}(a, \theta = \pi/2, \lambda)\, dN(a).$$

For the particle size distribution law given above, and by substituting $x = 2\pi a/\lambda$, we derive

$$\frac{d\sigma}{\Omega d} = \frac{C}{4}\left(\frac{\lambda}{2\pi}\right)^{3-k} \int x^{2-k} Q_{sca}\,(x)\, dx.$$

According to Van De Hulst (1959, p. 418) the integral converges with $3 < k < 7$, even with limits zero and infinity, and regardless of the form of $Q(x)$. The integral varies only slightly when λ ranges over the visible wavelengths. Thus the observed wavelength dependence of the scattering should be as λ^{3-k}. Atmospheric haze scatters with a wavelength dependence of $\lambda^{-\alpha}$ where α usually ranges from 1 to 2.

According to our observations the wavelength dependence of the cross section corresponds to a value of $\alpha = 2{\cdot}5 \pm 1{\cdot}1$, hence the particle size distribution

exponent is $k = 5{\cdot}5 \pm 1{\cdot}1$. Without more data concerning the optical properties of the particles we cannot determine the lower size limit. If we allow the upper limit of the particle size range, a_{max}, to be much greater than the lower limit, a_{min}, and if we assume totally reflecting particles, then we find from considerations of the observed total cross section and the maximum possible total volume that $a_{min} = 1{\cdot}5\ \mu$. It is likely, however, that considerable absorption occurs, in which case the lower radius limit would be less than $1{\cdot}5\ \mu$. A value of $\alpha = 2$ is typical of particles in the transition region between Rayleigh scattering ($\alpha = 4$) and large particle Mie scattering ($\alpha = 0$) which occurs for $2\pi a_{min} \sim \lambda$, or in our case $a_{min} \sim 0{\cdot}1\ \mu$. In either case therefore the observations of the particulate ring confirm the results of the laboratory experiments which showed that the vented material consists predominantly of particles in the size range $0{\cdot}1 < a < 1\ \mu$, the high degree of fragmentation being due to the inclusion of an azide in the chemical mix. The higher exponent k reported here is consistent with the available nitrogen being 2·6 times higher by mass than in the laboratory tests.

We find, therefore, that although the particle size distribution can be determined from the data, more quantitative data on absorption by the particle material is necessary to derive a value of the total particulate mass. If the particles were non-absorbing, the maximum available volume would result in a total scattered intensity several times the observed value.

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